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SUPERREVIVALS OF RYDBERG WAVE PACKETS[1]

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The revival structure and evolution of Rydberg wave packets are studied on a time scale much greater than the revival time $t_{\rm rev}$. We find a new level of revival structure and periodic motion different from that of the known fractional revivals. The new sequence of revivals culminates with the formation of a wave packet that more closely resembles the initial packet than does the full revival at time $t_{\rm rev}$. We refer to such a revival as a superrevival. We also show that an initial radial wave packet may be described as a type of squeezed state known as a radial squeezed state. Our results apply not only for hydrogenic wave packets, but for wave packets in alkali-metal atoms as well in the context of quantum defect theory.

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1. Introduction

If a Rydberg atom is excited by a short-pulsed laser field, a superposition of states with a spread of energy levels results [1]. Experiments on such systems have detected electronic motion with a periodicity $T_{\rm cl}$ equal to the classical period of a particle in a keplerian orbit. However, the motion is not entirely classical, as the wave packet disperses with time. After many Kepler orbits the wave packet recombines into nearly its original shape at the revival time $t_{\rm rev}$. Moreover, prior to this full revival, the wave function evolves through a sequence of fractional revivals, which consist of distinct subsidiary waves moving with a period that is a fraction of $T_{\rm cl}$ [1, 2, 3]. These fractional revivals have been observed in time-delayed photoionization and phase modulation experiments.

In the first part of this talk, we examine the revival structure and evolution of hydrogenic Rydberg wave packets for times much greater than the revival time [4]. We then consider the case of radial wave packets and show that the motion of these wave packets has features characteristic of squeezed states, and we outline an approach for a squeezed-state description [5]. In the final section, we describe how a quantum-defect theory based on supersymmetry may be used to model wave packets in alkali-metal atoms. In this context, we show that the dependence on the quantum defects of the

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long-term revival times for Rydberg wave packets in alkali-metal atoms is different from that of the laser detunings [6].

2. Superrevivals of Rydberg Wave Packets

The time-dependent wave function for a hydrogenic wave packet may be expanded in terms of energy eigenstates as

$$\Psi(\vec{r},t) = \sum_{n=1}^{\infty} c_n \varphi_n(\vec{r}) \exp\left[-iE_n t\right] \quad . \tag{1}$$

Here, $E_n = -1/2n^2$ is the energy in atomic units, and $\varphi_n(\vec{r})$ represents a generic form of the wave function. For a circular wave packet, $\varphi_n(\vec{r}) = \psi_{n,n-1,n-1}(\vec{r})$, whereas for a radial wave packet, $\varphi_n(\vec{r}) = \psi_{n,1,0}(\vec{r})$, where $\psi_{nlm}(\vec{r})$ is a hydrogen eigenstate of energy and angular momentum.

Both these types of wave packet are excited by a short laser pulse. The laser can be tuned to excite coherently a superposition of states centered on a mean value $\bar{n}$ of the principal quantum number. In what follows we assume that the distribution is strongly centered around $\bar{n}$. We may therefore approximate the square of the weighting coefficients c_n as a gaussian function of width σ.

If we expand the energy in a Taylor series around the centrally excited value $\bar{n}$, we find that the derivative terms define distinct time scales that depend on $\bar{n}$: $T_{\rm cl} = 2\pi/E'_{\bar{n}} = 2\pi\bar{n}^3$, $t_{\rm rev} = -2\pi/\frac{1}{2}E''_{\bar{n}} = \frac{2\bar{n}}{3}T_{\rm cl}$, and $t_{\rm sr} = 2\pi/\frac{1}{6}E'''_{\bar{n}} = \frac{3\bar{n}}{4}t_{\rm rev}$. The first time scale, $T_{\rm cl}$, is the *classical keplerian period*. It controls the initial behavior of the packet. The second time scale, $t_{\rm rev}$, is the *revival time*. It governs the appearance of fractional and full revivals. The third time scale, $t_{\rm sr} \gg t_{\rm rev}$, is a larger time scale we refer to as the *superrevival time*. This time scale determines the behavior of the packet for times greater than $t_{\rm rev}$.

Keeping terms through third order, and defining the integer index $k = n - \bar{n}$, we may write the wave function as

$$\Psi(\vec{r},t) = \sum_{k=-\infty}^{\infty} c_k \varphi_k(\vec{r}) \exp\left[-2\pi i\left(\frac{kt}{T_{\rm cl}} - \frac{k^2 t}{t_{\rm rev}} + \frac{k^3 t}{t_{\rm sr}}\right)\right] \quad . \tag{2}$$

We have found that at certain times $t_{\rm frac}$ it is possible to expand the wave function $\Psi(\vec{r},t)$ of Eq. (2) as a series of subsidiary wave functions. The idea is to express $\Psi(\vec{r},t)$ as a sum of wave functions $\psi_{\rm cl}$ with matching periodicities and with shape similar to that of the initial wave function $\Psi(\vec{r},0)$. We find that at certain times $t_{\rm frac} \approx \frac{1}{q}t_{\rm sr}$, where q must be an integer multiple of 3, the wave packet can be written as a sum of macroscopically distinct wave packets. Furthermore, at these times $t_{\rm frac}$, we also find that the motion of the wave packet is periodic with a period $T_{\rm frac} \approx \frac{3}{q}t_{\rm rev}$. Note

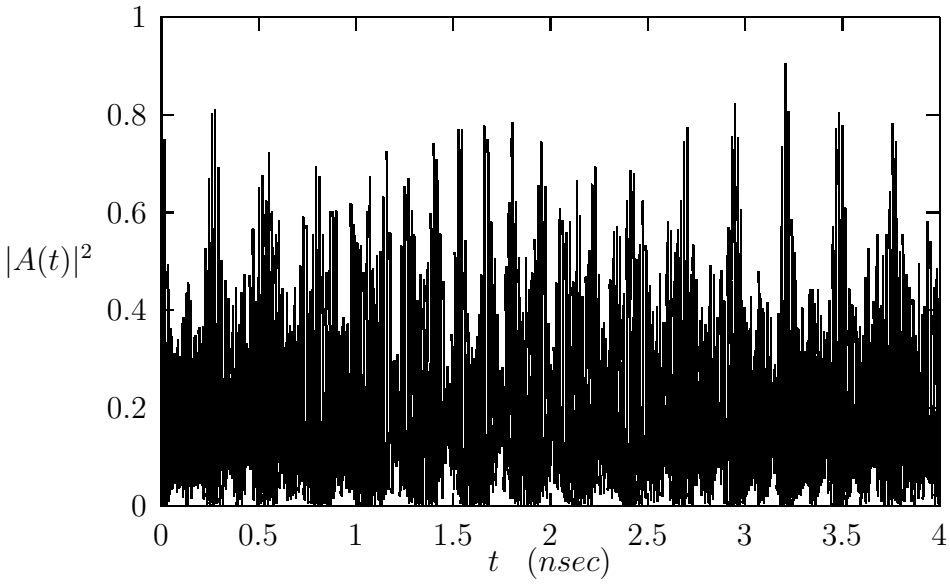

Figure 1: The absolute square of the autocorrelation function for a Rydberg wave packet with $\bar{n} = 48$ and $\sigma = 1.5$ is plotted as a function of time in nanoseconds.

that these periodicities are different from those of the fractional revivals, and thus a new level of revivals commences for $t > t_{\rm rev}$. We also find that at the particular time $t_{\rm frac} \approx \frac{1}{6} t_{\rm sr}$, a single wave packet forms that resembles the initial wave packet more closely than the full revival does at time $t_{\rm rev}$, i.e., a superrevival occurs.

In Refs. [4], we have given theoretical proofs for the periodicity and occurrence times of the superrevivals. We have looked at examples for large values of $\bar{n}$ that illustrate the structure of the fractional and full superrevivals clearly. This structure may be seen as well in examples with smaller values of $\bar{n}$. Figure 1 shows the square of the autocorrelation function for hydrogen with $\bar{n} = 48$ and $\sigma = 1.5$. In this case, the full revival is at $t \approx t_{\rm rev} \simeq 0.538$ nsec. For times greater than this, one can observe a fractional superrevival at $t \approx \frac{1}{12} t_{\rm sr} \simeq 1.61$ nsec with autocorrelation periodicity $T_{\rm frac} \approx \frac{1}{4} t_{\rm rev}$ and a full superrevival at $t \approx \frac{1}{6} t_{\rm sr} \simeq 3.23$ nsec with autocorrelation periodicity $T_{\rm frac} \approx \frac{1}{2} t_{\rm rev}$. The size of the peak in the autocorrelation function shows that the superrevival resembles the initial wave packet more closely than does the revival wave packet at $t \approx t_{\rm rev}$.

It appears likely that an experiment can be performed to detect the long-time effects described in this talk. One possibility is to use the pump-probe method of detection for Rydberg wave packets with $\bar{n} \approx 45 - 50$. This is experimentally feasible, provided a delay line of 3 – 4 nsec is installed in the apparatus. This should permit

detection of both full and fractional superrevivals.

3. Radial Squeezed States

In this part of the talk, we consider a description of radial Rydberg wave packets as a type of squeezed state. The theoretical analyses performed to date on radial wave packets rely on established tools such as expansion in eigenstates, numerical methods, perturbation theory, and/or the WKB approximation. However, the initial localization of the packet suggests it might be profitably described in terms of some kind of coherent state. Standard approaches along these lines either run into substantial technical difficulties or generate objects that do not match the behavior of p-state Rydberg atoms excited by short-pulsed lasers with no external fields.

To construct the squeezed states appropriate for a description of radial Rydberg wave packets, we have adopted a procedure used in Refs. [8] in the context of the construction of 'minimum-uncertainty coherent states.' The idea is to change variables from r and p_r to a new set, R and P, chosen to have oscillatory dependence on a suitable variable. The similarities between the ensuing equations and the usual quantum harmonic oscillator are sufficient to render possible an analytical construction of our candidate Rydberg wave packets. Our method generates a three-parameter family of radial squeezed states

$$\psi(r) = \frac{(2\gamma_0)^{2\alpha+3}}{\Gamma(2\alpha+3)} r^{\alpha} e^{-\gamma_0 r} e^{-i\gamma_1 r} \quad . \tag{3}$$

For purposes of comparison of our radial squeezed states with other theory and experiment, we determine the parameters α, γ_0, and γ_1 by fixing the form of the packet at the first pass through the classical apsidal point by the conditions $\langle p_r \rangle = 0$, $\langle r \rangle = r_{\rm out}$, $\langle H \rangle = E_{\bar{n}}$, where $r_{\rm out}$ is the outer apsidal point of the orbit and $E_{\bar{n}} = -1/2\bar{n}^2$ is the energy of the dominant state among those excited by the short laser pulse.

We have shown that these radial squeezed states may be used as an initial wave function to model the motion of a wave packet produced by a short laser pulse. The time evolution of the radial squeezed states exhibits the expected revival structure as well as the oscillations in the uncertainty product that are characteristic of a squeezed state.

4. Wave Packets in Alkali-Metal Atoms

All of the results described above for hydrogen may be rederived in the context of supersymmetry-based quantum-defect theory (SQDT) [7]. Since SQDT wave functions form a complete and orthonormal set with the correct eigenenergies for an

alkali-metal atom, the expansion of the energy for a Rydberg wave packet may be carried out in this context with the energies $E_{n^*} = -1/2n^{*2}$, where $n^* = n - \delta(l)$ and $\delta(l)$ is an asymptotic quantum defect for an alkali-metal atom. In this case, the Taylor expansion is carried out around a noninteger central value N^* that may or may not be on resonance. For the off-resonance case, the noninteger part of N^* consists of two parts: one from the quantum defect and another from the laser detuning. We may therefore consider quite generally the question of how the effects of quantum defects differ from those of a laser detuning in the evolution of the wave packet.

In Ref. [6], we have shown that the effects of the quantum defects are different from those of the laser detuning. This difference arises because a constant shift in the laser detuning is equivalent to a constant shift for all energy levels, whereas a constant shift in the quantum defect would correspond to varying shifts among the energy levels.

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